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Approximation for scattering properties of aggregated spherical particles: ERI method

S. JACQUIER¹, F. GRUY¹

¹Centre SPIN-Département GENRIC, UMR CNRS 5148 LPMG, Ecole des Mines de Saint-Etienne, 158, Cours Fauriel, 42023 Saint-Etienne Cedex 2 (France)

ABSTRACT

Powders of metallic oxides (ZrO₂, TiO₂, Al₂O₃, SiO₂ ...) are used as precursors in ceramic industry or as pigments in paints. Aggregation occurs in the suspension of such particles in water. The most often, the formed aggregates contain few primary particles. The on-line characterization of the suspension is made by spectral turbidimetry, i.e attenuation of a light beam by particles scattering and absorption. In order to analyse the corresponding signal, i.e turbidity, the scattering cross section and anisotropy factor of aggregates are needed. It exists an important literature about the calculation of the light scattering of aggregates. For instance, Xu [1] proposed an exact calculation based on a generalization of the Mie theory. In a previous work [2] we compare the exact value of scattering cross section (C_{sca}) got from Xu's theory to the corresponding one from the following approximations: Effective Refractive Index (ERI) method, Percival-Berry-Khlebtsov (PBK) method, Anomalous Diffraction (AD) approximation for different aggregates morphology. We conclude that ERI method is the most conclusive over all the size parameter range $\alpha \in [0.01; 10]$ and for the 3 materials. Tested materials are the ones with low (SiO₂, $m=1.08$), intermediate (Al₂O₃, $m=1.32$) and high (TiO₂, $m=1.94$) optical contrast in water. In turbidimetry, another relevant parameter is the radiation pressure cross section (C_{pr}). The topic of this paper is, to extend this work by comparing the C_{pr} obtained with the exact theory and the one calculated with ERI method.

1. INTRODUCTION

Many industrial processes involve solid-liquid suspensions (eg: paintings). These suspensions, initially made by solid primary particles, contain many aggregates which modify their properties (eg. the opacity). The characterization methods (eg. size of primary particle) of these suspensions use the light scattering (Mie's theory). However, Mie's theory (1908), which is used by the traditional granulometer, is only applicable to spherical primary particles and does not make possible to measure the aggregates. An extension of this theory to the case of the multiple scattering was given by Xu (1995-2003) ([1] for more information see the references therein): GMM (Generalized Multiparticle Mie solution). But the computation times of the optical properties via the exact theory, for aggregates, does not allow considering its use in-situ, in the immediate future. Our work, thus, directed towards the search for approximated methods for the optical properties of aggregated non-absorbent spherical particles [2][3][4]. Initially we evaluated with respect to the exact method, seven approximated methods allowing to obtain the scattering cross section.

An aggregate is composed of N primary spherical particles. The radius and the dimensionless size parameter of the primary particles are respectively denoted a_1 and $\alpha = \frac{2\pi}{\lambda} a_1$ (λ is the wavelength of the incident light). For a given number of primary particles different aggregates shape or configuration can be considered.

We concluded ([3], [4]) that:

- methods of the Compact Sphere (CS) and Hollowing Sphere (HS) are inappropriate

- methods using a fractal dimension are as for them not very conclusive on aggregates containing a low number of primary particles.

- Percival-Berry-Khlebtsov (PBK) method is valid for $0 < \alpha < 2$, with an error which increases with the refractive index of material.

- Anomalous Diffraction (AD) method is correct for $2 < \alpha < 10$, and is less sensitive to the increase of refractive index.

- Effective Refractive Index (ERI) is the approximated method being able to be considered on the whole size parameter.

In this article, we study more in detail this method and extend the comparison with respect to the exact method to the case of the radiation pressure cross section.

2. ERI METHOD

We will show in the part devoted to the optical characterization of an aggregate with the exact method (part 3), that the location of the primary particles within the aggregate has an influence on the cross sections. The ERI method takes into account of the morphology because we consider an equivalent sphere defined from the aggregates projected surface.

The principle is as follows. The aggregate is projected for several successive plans (considered perpendicular to the incident wave vector), this represents a mobile aggregate which would undergo a random rotation in a fluid. From each projection, a surface (S_p) is determined, and following these multiple projections, an average surface ($\langle S_p \rangle$) is obtained which represents an equivalent sphere projected area.

$$\pi a_{N,e}^2 = \langle S_p \rangle_O$$

We can, thus, deduce, successively, an equivalent diameter ($2a_{N,e}$), a volume fraction of solid in the sphere (Φ_a), an effective relative refractive index (m_a) with the Maxwell-Garnett theory and then, the cross sections with the Mie's theory.

$$\bar{\phi}_a = (Na_1^3)/a_{N,e}^3$$

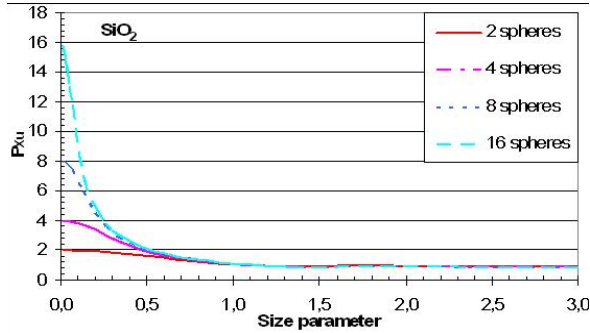
$$\frac{m_a^2 - 1}{m_a^2 + 2} = \bar{\phi}_a \frac{m^2 - 1}{m^2 + 2}$$

3. EXACT METHOD

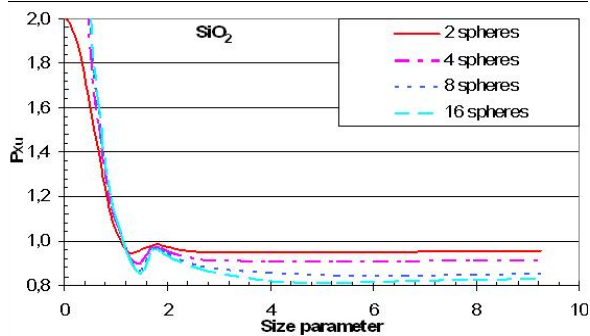
Here, only the results for C_{pr} in the case of a linear configuration are showed (Figure 1), those relative to C_{sca} are showed in [2],[3] and [4].

P_{Xu} is defined as the ratio of the radiation pressure cross section of an aggregate by the sum of radiation pressure cross section of N primary particles which form the aggregate (so, the denominator considers the case of non interacting spheres).

$$P_{Xu} = \frac{C_{Xu,N}}{NC_{Mie,1}} \quad (1)$$



a)



b)

Figure 1: P_{Xu} as function of the size parameter for a linear configuration with N primary particles for SiO_2
a: $0 < P_{Xu} < 18$, b: $0.8 < P_{Xu} < 2$

The study of the parameters influencing scattering (C_{sca}) and the radiation pressure (C_{pr}) cross sections of aggregates obtained with the exact method revealed that:

- various configurations of an aggregate following its shape or the number of primary particles which it contains are perfectly distinguishable,

- there are two α -ranges. One, where the cross sections have values largely higher than, the one of an identical group of particles without interaction, the other, where they are on the contrary lower.

- the number of primary particles is the relevant parameter for small size parameter ($\alpha \rightarrow 0, C_{Xu,N} \propto N^2 C_{Mie,1}$),

- there are two extreme configurations (linear and compact) between which the cross sections of the others evolve,

- the difference of the cross sections of the two extreme configurations increases with the number of primary particles.

In the second range $\alpha > 1.5$ (figure 1), the ratio P_{Xu} tends towards a pseudo-constant.

4. APPROXIMATION FOR $\alpha > 1.5$

In this part we determine the proportionality factor between $C_{pr Xu, N}$ and $C_{pr Xu, 1}$ or $C_{pr Xu, 2}$

We have noticed in the exact method study that P_{Xu} was, for α higher than 1.5, constant for an aggregate in a given configuration. This constant, denoted p , is also a function of the material refractive index (SiO_2 , Al_2O_3 , TiO_2). Its dependence on the aggregate morphology can be expressed by using N and d_1 (the mean inter-primary particle distance(2)).

$$d_1 = \frac{1}{N(N-1)} \sum_{i \neq j} d_{i,j} \quad (2)$$

$d_{i,j}$ is the distance between primary particles i and j

We will successively examine two approximate methods:

➤ $C_{pr Xu, N}$ proportional to C_{pr} of a primary particle: $C_{pr Mie, 1}$

We evaluated p , which we call p_1 , according to the material refractive index and aggregate morphology.

This leads us to express p_1 as follows:

- for SiO_2 : $p_1 = 1 - 0.027(\beta)^{0.8}$

- for Al_2O_3 : $p_1 = 1 - 0.056(\beta)^{0.7}$

- for TiO_2 : $p_1 = 1 - 0.1(\beta)^{0.65}$

Thus, we define: $P_{p1} = \frac{p_1 NC_{pr Mie, 1}}{C_{pr Xu, N}}$

➤ $C_{pr Xu, N}$ proportional to C_{pr} of a doublet: $C_{pr Xu, 2}$

We evaluated p , which we call p_2 , according to the material refractive index and aggregate morphology. Then the following expressions are obtained:

- for SiO_2 : $p_2 = 1 - 0.021(\beta)^{0.9}$

- for Al_2O_3 : $p_2 = 1 - 0.08(\beta)^{0.51}$

- for TiO₂: $p_2 = 1 - 0.07(\beta)^{0.8}$

Thus, we define: $Pp_2 = \frac{p_2(N/2)C_{prMie,2}}{C_{prXu,N}}$

p_1 and p_2 are a function of N and d_1 : $\beta = \frac{N}{\sqrt{d_1}/2}$

We will test this two approximations at the end of the next part.

5. ERI PERFORMANCE

In the same way that we showed the effectiveness of the approximated method ERI for calculation of C_{sca} , we will evaluate his performances for the calculation of C_{pr} . We consider the ratio R_m of the radiation pressure cross sections obtained with the ERI method and with the exact method:

$$R_m = \frac{C_{ERI,N}}{C_{Xu,N}} \quad (3)$$

On the Figure 2 is presented (only for SiO₂) the change of this ratio, according to the size parameter of the primary particles constituting the aggregate. C_{pr} is calculated for various types of aggregates (compact or linear configuration) made up with 2, 4, 8, 16 primary particles. Let us recall that if the approximate method is perfect, R_m is equal to 1. Some is the configuration or the material; the curves shape is similar to those obtained for calculation of C_{sca} ([3], [4]). Indeed, in the case of the linear configurations, we also notice the presence of a peak for size parameters lower than 2. This behaviour was awaited, because the asymmetry parameter tends towards 0, when α tends towards 0, i.e. scattering and the radiation pressure cross sections become equal. For $\alpha > 2$, the approximate method also deviates from the exact method.

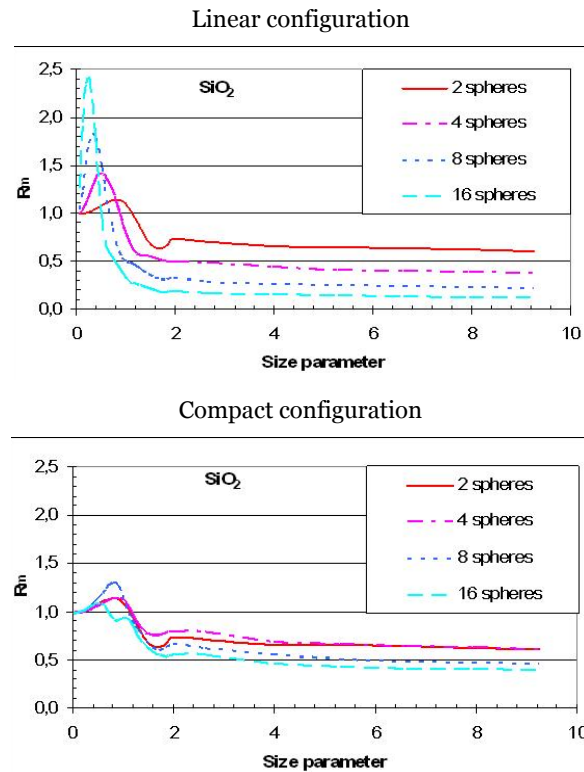


Figure 2: ERI Method: R_m for the radiation pressure cross section, for α ranging between 0 and 10 for SiO₂ material and for linear and compact configurations

		0< α <2											
		Linear configuration						Compact configuration					
index	SiO ₂		Al ₂ O ₃		TiO ₂		SiO ₂		Al ₂ O ₃		TiO ₂		
method	m	σ	m	σ	m	σ	m	σ	m	σ	m	σ	
ERI	0,91	0,47	0,85	0,44	0,78	0,39	0,97	0,17	0,93	0,15	0,83	0,30	
ERI/G	1,29	0,39	1,16	0,25	0,94	0,13	1,06	0,14	1,02	0,11	0,87	0,29	

Table 1: Performance for the calculation of C_{pr} for different material with $\alpha \in [0; 2]$ with ERI and ERI corrected (ERI/G) method.

The Table 1 and 2 gather the performances of ERI method for the two α -ranges.

2< α <10												
Linear configuration							Compact configuration					
index	SiO ₂		Al ₂ O ₃		TiO ₂		SiO ₂		Al ₂ O ₃		TiO ₂	
method	m	σ	m	σ	m	σ	m	σ	m	σ	m	σ
ERI	0,36	0,66	0,38	0,65	0,55	0,49	0,57	0,44	0,60	0,41	0,76	0,30
ERI/G	0,93	0,17	0,90	0,15	1,07	0,23	0,92	0,13	0,90	0,11	1,06	0,30

Table 2: Performance for the calculation of C_{pr} for different material with $\alpha \in [2; 10]$ with ERI and ERI corrected (ERI/G) method.

The average (m) and the standard deviation (σ) are calculated from $C_{pr}(\alpha, N)$ values for each configuration and material.

ERI method for the calculation of C_{pr} is not very effective for size parameter higher than 2, because even in the case of the compact configurations (the average effectiveness is 0.97 for the scattering cross section) the average effectiveness is about 0.64.

These considerations lead us to express the function $G(\alpha)$, making possible to correct C_{pr} obtained with the approximate method ERI. Firstly, we express the maximum value of α (see figure 2) as:

$$\alpha_{\max} \approx 1 / 2.3(\gamma)^{0.92}, \gamma = d_1 / 2\sqrt{N}$$

Then,

If $\alpha < \alpha_{\max}$ $G = F_2$

else,

$$G = F_2 + (1 - h_{\min}) \left(e^{-2 \left(\frac{\alpha}{\alpha_{\max}} - 1 \right)} - 1 \right)$$

where F_2 is defined in [4] by:

$$F_2 = 1 + R_{\max} \left(\frac{\alpha}{\alpha_{\max}} \right)^n e^{-2n \left(\frac{\alpha}{\alpha_{\max}} - \sqrt{\frac{\alpha}{\alpha_{\max}}} \right)} \quad \text{with}$$

$$R_{\max} = 0.33(1/\gamma)^{-2.1} \text{ and } n = 0.5e^{2/\gamma}$$

$$h_{\min} = 0.1 \ln(8N^{0.1}/d_1^3) + 0.7$$

F_2 , n , R_{\max} and α_{\max} were initially defined to correct C_{sca} obtained with ERI method. F_2 is a gamma type function which is elaborated to correct C_{sca} in the case of low size parameter ($\alpha < 2$).

Thus, G is built in the same framework with

The corrected radiation pressure cross section will obey the expression: $C_{pr,ERI,N}^C = C_{pr,ERI,N} / G$

The tables 1 and 2 contain the data for ERI method corrected by G function. ERI method is improved with the use of the function G .

The data in table 3, show that the ERI/ G method is not as efficient as Pp1 and Pp2. But ERI/ G has the biggest advantage to be used over all the size range.

1,5 < α < 10												
Linear configuration							Compact configuration					
index	SiO ₂		Al ₂ O ₃		TiO ₂		SiO ₂		Al ₂ O ₃		TiO ₂	
method	m	σ	m	σ	m	σ	m	σ	m	σ	m	σ
Pp1	1,01	0,05	1,05	0,11	1,06	0,17	1,01	0,04	1,00	0,11	1,08	0,17
Pp2	0,98	0,04	0,96	0,08	0,94	0,12	0,97	0,04	0,93	0,10	0,93	0,11
ERI/G	0,93	0,17	0,90	0,15	1,07	0,23	0,92	0,13	0,90	0,11	1,06	0,30

Table 3: Approximated method performance for the calculation of C_{pr} for different material with $\alpha \in [1.5; 10]$.

6. CONCLUSION

C_{pr} and C_{sca} for an aggregat behave in the same way over the primary particle size parameter range. We obtain satisfactory results with ERI method. The use of the correction functions F and G makes possible to improve considerably ERI method. Further, the modification of these correction functions, in the aim to increase the performances, will be the subject of future investigations.

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